

## SHORT COMMUNICATIONS

### Phosphorus Flux from Bottom Sediments in Lake Eucha, Oklahoma

B. E. Haggard,\* P. A. Moore, Jr., and P. B. DeLaune

#### Abstract

Phosphorus inputs into reservoirs include external sources from the watershed and internal sources from the reservoir bottom sediments. This study quantified sediment P flux in Lake Eucha, northeastern Oklahoma, USA, and evaluated the effectiveness of chemical treatment to reduce sediment P flux. Six intact sediment–water columns were collected from three sites in Lake Eucha near the reservoir channel at depths of 10 to 15 m. Three intact sediment and water columns from each site were incubated for 21 d at approximately 22°C under aerobic conditions, and three were incubated under anaerobic conditions (N<sub>2</sub> with 300 ppm CO<sub>2</sub>); sediment P flux was estimated over the 21 d for each core. The overlying water in the cores was bubbled with air for approximately 1 wk and then treated with aluminum sulfate (alum). The cores were incubated at approximately 22°C for an additional 14 d under aerobic or anaerobic conditions, and sediment P flux after alum treatment was estimated for each core. Sediment P flux was approximately four times greater under anaerobic conditions compared with aerobic conditions. Alum treatment of the intact sediment–water columns reduced (8×) sediment P flux under anaerobic conditions. Internal P flux (1.03 and 4.40 mg m<sup>-2</sup> d<sup>-1</sup> under aerobic and anaerobic conditions, respectively) was greater than external P flux (0.13 mg m<sup>-2</sup> d<sup>-1</sup>). The internal P load (12 Mg yr<sup>-1</sup>) from reservoir bottom sediments was almost 25% of the external P load (approximately 48 Mg yr<sup>-1</sup>) estimated using a calibrated watershed model.

PHOSPHORUS FLUX from sediments in many lakes and reservoirs are sometimes sufficient to maintain anthropogenic eutrophication when external P sources have been reduced (Larsen et al., 1979; Ryding, 1981). Under some circumstances, high P loads from external sources may eventually provide an internal P source from bottom sediments to the overlying water column when the hypolimnion becomes anoxic in stratified lakes and reservoirs. Under anaerobic conditions at the sediment–water interface, P may be released when Mn and Fe minerals are subsequently reduced (Mortimer, 1941, 1942). Anaerobic P release from sediments is dependent on many factors including Eh and pH (Anderson, 1975; Moore and Reddy, 1994), P forms in sediments (James

et al., 1995, 2000), and temperature and microbial activity (Riley and Prepas, 1984; Gächter et al., 1988).

Several studies have evaluated the ability of sediments to release P under aerobic and anaerobic conditions using intact sediment–water cores (e.g., see Anderson, 1975; Holdren and Armstrong, 1980; Moore and Reddy, 1994; Moore et al., 1998). Sediment P flux was generally much greater under anaerobic conditions than under aerobic conditions, probably related to oxidation–reduction interactions between adsorbed P and Fe and Mn minerals. Mineral solubility may also control P concentrations near the sediment–water interface or in pore water, thus regulating sediment P flux to the overlying water column. For example, equilibrium calculations at the sediment–water interface have shown P solubility to be controlled by Fe mineral precipitation under aerobic conditions and by Ca mineral precipitation under anaerobic conditions (Moore et al., 1991, 1998). In calcareous systems, P release may be related to Ca–P mineral solubility in pore waters and may be much less dependent on the oxidation status of overlying water (Golterman, 1982; Moore et al., 1991).

The treatment of bottom sediments with chemical amendments [e.g., aluminum sulfate, (Al<sub>2</sub>SO<sub>4</sub>)<sub>3</sub>, and gypsum, CaSO<sub>4</sub>, etc.] may be a successful means to ameliorate high sediment P fluxes (Kennedy and Cooke, 1982; Welch and Schriever, 1994; Varjo et al., 2003). Alum treatment to lakes and reservoirs is an effective, economical method to improve water quality where added alum forms an Al hydroxide floc that removes P and colloidal organic matter from the water column (Francko and Heath, 1981; Kennedy et al., 1987). Several studies have observed significant reductions in water column total P concentration, chlorophyll *a* concentrations, and the abundance of blue-green algae after lakes or reservoirs have received alum treatment (e.g., see Kennedy et al., 1987; Welch and Schriever, 1994). However, Hullebusch et al. (2002) showed that alum treatment had variable effects on water quality and the occurrence of blue-green algae (*Microcystis* sp.) did not cease after alum treatment.

The purpose of this study was to: (i) quantify internal P flux from reservoir bottom sediments in Lake Eucha, Oklahoma, April 2002, using intact sediment and water cores incubated under aerobic and anaerobic conditions, and (ii) evaluate the effectiveness of alum in reducing sediment P fluxes under aerobic and anaerobic conditions. This paper also compared internal P flux estimated using intact sediment and water cores to external

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**Abbreviations:** SRP, soluble reactive phosphorus.

P flux estimated using a watershed hydrologic and water quality model in the discussion.

## Materials and Methods

### Study Site Description

The Lakes Eucha and Spavinaw system is the municipal drinking water supply to almost 500 000 people in the greater Tulsa metropolitan area and also smaller communities surrounding these reservoirs in northeastern Oklahoma; Lake Eucha is the upstream reservoir in this two-reservoir system. Recently, these drinking water supply reservoirs have had taste and odor problems in finished waters resulting from the prevalence of some blue-green algae and diatoms. These water quality problems and a doubling in reservoir total P concentrations from 1975 to 1995 (Wagner and Woodruff, 1997) have prompted several other studies investigating P loads and retention from external sources in the catchment.

A comprehensive hydrologic and watershed model has been calibrated for this catchment (Storm et al., 2002); the model used was the Soil and Water Assessment Tool (SWAT). The model was calibrated using stream water quality monitoring data and discharge from 1998 through 2001. Measured stream discharge was limited to three U.S. Geological Survey stream gages in the basin whereas water-quality samples were collected at eight sites. The model used: (i) topography defined by a Digital Elevation Model (DEM); (ii) soil data from the State Soil Geographic Database (STATSGO); (iii) land cover that was developed by satellite imagery and ground-truthed specifically for this project; and (iv) weather data where daily rainfall estimates with greater spatial resolution were derived from Next Generation Weather Radar (NEXRAD). The model included 62 subbasins and 1052 hydraulic response units (HRUs). The model estimated average annual P loads at approximately 48 Mg yr<sup>-1</sup> (1998–2001) from the 932-km<sup>2</sup> catchment draining into Lake Eucha. This estimated load includes 11.4 Mg yr<sup>-1</sup> from the City of Decatur wastewater treatment plant (Storm et al., 2002).

The current study focused on internal P sources where three sediment sampling sites were chosen in April 2002 (Site 1: 36°21.032' N, 94°49.195' W; Site 2: 36°20.774' N, 94°52.580' W; Site 3: 36°21.615' N, 94°53.459' W). Site 1 was in the upper or riverine reaches of Lake Eucha whereas Sites 2 and 3 were in the transitional zone of this small reservoir (Fig. 1); Site 3 is the most down-reservoir sediment sampling station. All three sites had typical mud sediments, often referred to as *gyttja*.

### Sediment Phosphorus Flux

Six intact sediment–water columns (hereafter, cores) were collected at each sediment sampling site using a certified scuba diver and 1-m Plexiglas tubes (6.35-cm inside diameter). The tubes were inserted into the sediments approximately 0.5 m, and then carefully removed from the sediments and closed at each end using #13 rubber stoppers. A properly collected core had relatively undisturbed sediment at the surface and with depth, and the overlying water in the core was not turbid from sediment disturbance but relatively clear. Upon return to the laboratory, the depth of overlying reservoir water in the sediment–water cores was adjusted, so that each core contained 1 L of overlying water. The cores were wrapped in Al foil to exclude light and incubated at room temperature, approximately 22°C. Three cores were incubated under aerobic condi-

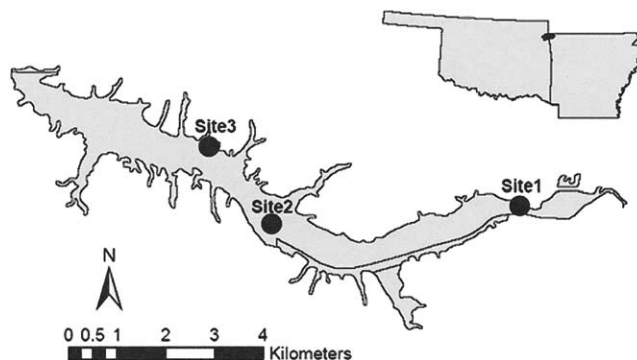


Fig. 1. Map showing the Lake Eucha catchment in Arkansas and Oklahoma and the three sediment sampling sites at Lake Eucha, Oklahoma.

tions and three under anaerobic conditions from each site. The aerobic sediment–water cores had air bubbled through the water column whereas the anaerobic sediment–water cores had nitrogen gas (N<sub>2</sub>) with 300 ppm CO<sub>2</sub> bubbled through the water column. The cores were incubated for more than 21 d under these (aerobic and anaerobic) conditions and then the water column of all cores was bubbled with air for approximately 1 wk. Approximately 0.25 g of solid alum [Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>·14H<sub>2</sub>O] was added directly to the overlying water in the sediment–water cores, representing an application rate of approximately 79 g m<sup>-2</sup>. A floc was formed by the alum additions that settled to the sediment surface, and the treated cores were incubated again for 14 d under aerobic or anaerobic conditions. This rate (0.25 g core<sup>-1</sup>) of alum treatment equates to approximately 79 g m<sup>-2</sup>, or a >900-Mg alum treatment to Lake Eucha.

At 1- to 3-d intervals during the incubation periods, a 60-mL water sample was removed from the overlying water in the cores. This water sample was used to measure pH and conductivity, and a portion was filtered through 0.45-μm membrane, acidified using concentrated HCl, and analyzed for soluble reactive phosphorus (SRP) using the automated ascorbic acid reduction technique (American Public Health Association, 1998). Soluble metals (Al, Ca, Fe, K, Mg, Mn, Mo, Na, S, and Zn) were also determined on this filtered, acidified water sample using inductively coupled argon plasma (ICAP) with a Model D ICP (Spectro Analytical Instruments, Fitchburg, MA). The overlying water was maintained at a volume of 1 L using filtered (0.45-μm membrane) reservoir water that was collected at Lake Eucha with a known (i.e., measured) SRP concentration.

Sediment P fluxes were calculated as linear changes in P mass in the overlying water (after corrections for the 60-mL removal of overlying water in the core and with replacement of reservoir water) as a function of time divided by the inside area of the sediment–water cores. Simple linear regression was used to determine the slope of the relation between adjusted SRP mass and time. This slope represented the linear change in P mass with time (mg d<sup>-1</sup>) and was divided by the core area (0.0032 m<sup>2</sup>) to determine sediment P flux (mg m<sup>-2</sup> d<sup>-1</sup>). Specific comparisons made in this study were (Objective 1) sediment P flux during aerobic and anaerobic conditions and (Objective 2) sediment P flux before and after alum treatment of the water column. Because the SRP concentration in the overlying water was increasing with time, the concentration gradient for SRP would decrease with time; hence, the flux estimates reported herein are probably somewhat less than what may actually occur in Lake Eucha (if sorption process are the primary mechanism for P release). The role of microbial

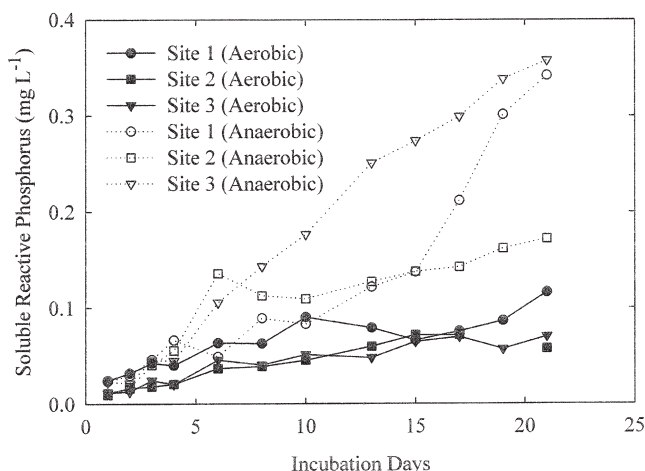


Fig. 2. Soluble reactive phosphorus concentrations under aerobic and anaerobic conditions with incubation time in the overlying water of the intact sediment cores from three sites in Lake Eucha (symbols are the mean of three data points).

mobilization of P may also be important over the total 42-d incubation period (Gächter et al., 1988).

## Results

Average SRP concentration of the filtered replacement water was approximately  $0.01 \text{ mg L}^{-1}$ . Initial SRP concentrations in the overlying water were greater than  $0.02 \text{ mg L}^{-1}$  in the sediment–water cores from Site 1, whereas SRP concentrations were approximately  $0.01 \text{ mg L}^{-1}$  in the cores from Sites 2 and 3. The SRP concentrations in the overlying water increased with time in all cores, although the rate of increase was greater under anaerobic conditions compared with aerobic conditions at all three sites (Fig. 2). After 21 d of incubation, the final SRP concentration in the overlying water of the cores from Sites 1 to 3 was between  $0.05$  and  $0.10 \text{ mg L}^{-1}$  under aerobic conditions and  $0.17$  and  $0.36 \text{ mg L}^{-1}$  under anaerobic conditions. The SRP concentrations in the overlying water were significantly correlated with soluble Mo, S, and Zn concentrations under aerobic conditions and soluble Mn, Mo, S, and Zn concentrations under anaerobic conditions (Table 1).

Table 1. Correlation between soluble reactive phosphorus (SRP) and soluble metal concentrations under aerobic and anaerobic conditions in the overlying water of the intact sediment cores before and after alum treatment at three sites in Lake Eucha, Oklahoma, April 2002.

Element	Pearson's correlation coefficient with SRP			
	Before 0.25-g alum dose ( $n = 36$ )		After 0.25-g alum dose ( $n = 27$ )	
	Aerobic	Anaerobic	Aerobic	Anaerobic
Al	−0.02	−0.19	−0.25	−0.28
Ca	0.12	−0.17	0.35	0.41*
Fe	−0.22	0.06	−0.12	0.24
K	0.13	0.16	−0.06	0.45*
Mg	0.13	−0.09	0.41*	0.33
Mn	0.29	0.46*	−0.16	0.38*
Mo	−0.55*	−0.51*	−0.09	−0.04
Na	−0.08	−0.24	−0.36	0.45*
S	−0.42*	−0.71*	−0.41*	0.05
Zn	−0.33*	−0.39*	0.15	0.25

\* Significant at the 0.05 probability level.

Table 2. Release rates of soluble reactive phosphorus (SRP) from reservoir bottom sediments using change in phosphorus mass in the overlying water of intact sediment cores versus time before and after alum treatment at three sites in Lake Eucha, Oklahoma, April 2002.

Sediment sampling station	Sediment P flux	
	Aerobic	Anaerobic
	$\text{mg m}^{-2} \text{ d}^{-1}$	
	Before 0.25-g alum dose	
Site 1	1.14	4.70*
Site 2	1.01	2.46*
Site 3	0.95	6.05*
Mean	1.03	4.40
	After 0.25-g alum dose	
Site 1	1.72	0.58*
Site 2	0.91	0.54
Site 3	0.71	0.53
Mean	1.11	0.55

\* Significant differences at the 0.05 probability level between the slopes of the relations between time and SRP concentrations used to estimate sediment P flux at each site under aerobic and anaerobic conditions.

The changes in SRP concentrations with time in the overlying water of the cores from Sites 1 to 3 produced significantly different estimates of sediment P flux under aerobic and anaerobic conditions (Table 2). The slope of the relation used to estimate sediment P flux was significantly greater at all sites under anaerobic conditions compared with aerobic conditions (Site 1:  $F = 35.3$ ,  $P < 0.0001$ ; Site 2:  $F = 12.7$ ,  $P = 0.002$ ; Site 3:  $F = 635.9$ ,  $P < 0.0001$ ). Sediment P flux was similar ( $0.95$ – $1.14 \text{ mg m}^{-2} \text{ d}^{-1}$ ) under aerobic conditions between Sites 1 to 3 (slope comparison,  $F = 0.4$ ,  $P = 0.65$ ). However, sediment P flux was significantly different under anaerobic conditions at each site (slope comparison,  $F = 20.6$ ,  $P < 0.0001$ ), ranging from the least at Site 2 ( $2.46 \text{ mg m}^{-2} \text{ d}^{-1}$ ) to the greatest at Site 3 ( $6.05 \text{ mg m}^{-2} \text{ d}^{-1}$ ). Average sediment P flux under aerobic and anaerobic conditions was  $1.03$  and  $4.40 \text{ mg m}^{-2} \text{ d}^{-1}$  from the sediment–water cores collected at Lake Eucha.

After alum treatment, initial SRP concentrations generally decreased from  $0.01$  to  $0.02 \text{ mg L}^{-1}$  to generally less than  $0.01 \text{ mg L}^{-1}$  the following day (Fig. 3). Average

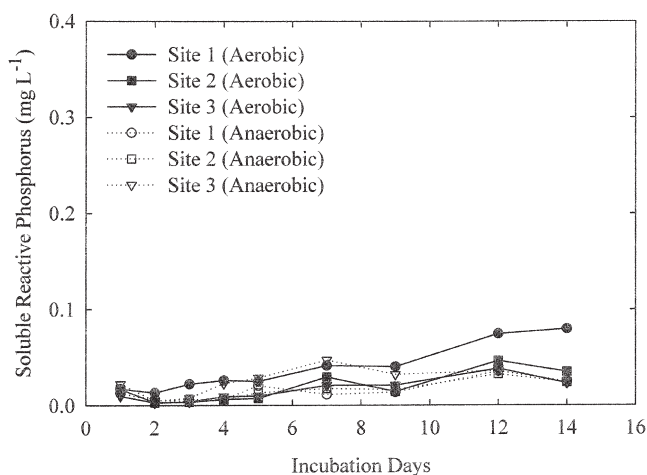


Fig. 3. Soluble reactive phosphorus concentrations following alum treatment under aerobic and anaerobic conditions with incubation time in the overlying water of the intact sediment cores from three sites in Lake Eucha (symbols are the mean of three data points).



SRP concentrations then generally increased from approximately  $0.01 \text{ mg L}^{-1}$  to between  $0.02$  and  $0.04 \text{ mg L}^{-1}$ , although one set of cores (from Site 1) increased to approximately  $0.08 \text{ mg L}^{-1}$  under aerobic conditions. After alum treatment, SRP concentrations in the overlying water of the intact sediment–water cores were significantly correlated with soluble Mg and S concentrations under aerobic and soluble Ca, K, Mn, and Na concentrations under anaerobic conditions (Table 1).

The slope of the relation used to estimate sediment P flux under anaerobic conditions was significantly less at all sites after alum treatment of the overlying water in the cores (Table 2; Site 1:  $F = 20.6$ ,  $P = 0.003$ ; Site 2:  $F = 11.5$ ,  $P = 0.004$ ; Site 3:  $F = 254.0$ ,  $P < 0.0001$ ). Sediment P flux during aerobic conditions was similar before and after alum addition at Sites 2 and 3 whereas sediment P flux was greater after alum treatment at Site 1 (slope comparison,  $F = 25.1$ ,  $P = 0.0002$ ). Sediment P flux under anaerobic conditions was similar across sites after alum treatment, whereas sediment P flux under aerobic conditions was greatest at Site 1 following alum addition to the overlying water of the cores (slope comparison,  $F = 7.9$ ,  $P = 0.003$ ).

## Discussion

Hypolimnetic P concentrations are probably dependent on mineral solubility, sediment sorption processes, and microbial mineralization during oxic conditions and redox potential during anoxic conditions. When the hypolimnion becomes anoxic, facultative anaerobic bacteria near the sediment–water interface use alternative electron acceptors, often reducing Fe and Mn minerals thereby releasing sorbed P (Mortimer, 1941, 1942). However, SRP concentrations in the overlying water of the sediment–water cores from Lake Eucha were not significantly correlated to soluble Fe concentrations during aerobic or anaerobic conditions. The lack of a relation between SRP and soluble Fe concentrations was probably because siderite ( $\text{FeCO}_3$ ) may control Fe solubility and limit soluble Fe concentration to less than  $0.1 \text{ mg L}^{-1}$  (Moore and Reddy, 1994). Soluble Mn concentrations were significantly correlated to SRP concentrations suggesting Mn reduction may have some role in the release of P from reservoir bottom sediments to the water column. Sediment P flux under aerobic and anaerobic conditions is also often correlated to interstitial P, loosely bound P, and NaOH-extractable P when sequential P extractions are used on reservoir bottom sediments (James et al., 1995, 2000).

This study clearly showed the importance of anaerobic conditions during internal P cycling within Lake Eucha, that is, when P is released from reservoir bottom sediments into the water column. In Lake Eucha, sediment P flux under anaerobic conditions was more than four times greater than that measured during aerobic conditions using the sediment–water cores. However, some spatial variability in anaerobic sediment P release rates was observed at Lake Eucha. Overall, sediment P flux under aerobic and anaerobic conditions was within the range ( $1\text{--}15 \text{ mg m}^{-2} \text{ d}^{-1}$ ) reported for other eutro-

phic lakes and reservoirs across North America (e.g., see James et al., 1995, 2000; Moore and Reddy, 1994; Moore et al., 1991, 1998; Riley and Prepas, 1984).

Sediment P flux estimated using hypolimnetic P concentrations in Lake Eucha was  $2.39$  and  $2.86 \text{ mg m}^{-2} \text{ d}^{-1}$  when the reservoir was stratified in 1998 and 1999, respectively (Oklahoma Water Resources Board, 2001). These Oklahoma Water Resources Board estimates were within the range of sediment P release observed under anaerobic conditions in the current study ( $2.46\text{--}6.05 \text{ mg m}^{-2} \text{ d}^{-1}$ ). The Oklahoma Water Resources Board estimated an internal P load to the epilimnion of approximately  $2.5 \text{ Mg yr}^{-1}$  using equations from Chapra and Reckhow (1983). Epilimnetic internal P loading can be low because thermal stratification limits mixing between the epilimnion and the anoxic, nutrient-rich hypolimnion. However, when thermally stratified reservoirs undergo fall turnover, total internal P loading can be substantial and influence reservoir water quality. The current study estimated average sediment P flux at  $1.03$  and  $4.40 \text{ mg m}^{-2} \text{ d}^{-1}$  under aerobic and anaerobic conditions, respectively, resulting in a total internal P load of  $12 \text{ Mg yr}^{-1}$  (assuming a lake surface area of  $11.7 \text{ km}^2$  and an anoxic hypolimnion  $0.5 \text{ yr}$ ). Overall, sediment P release rates resulted in an internal P load that was approximately 25% of the  $48 \text{ Mg yr}^{-1}$  external P load reported by Storm et al. (2002), but sediment P release on an aerial basis was much greater than the external flux ( $0.13 \text{ mg m}^{-2} \text{ d}^{-1}$ ) from this  $932\text{-km}^2$  catchment.

After watershed management strategies reduce external P flux, reservoir recovery can be delayed many years if mechanisms controlling internal P flux are not included in restoration plans (Larsen et al., 1979; Ryding, 1981). The best method to reduce sediment P flux may be the addition of alum, where the Al floc binds dissolved P and would not be subject to oxidation–reduction reactions. Alum dosage rates should be based on internal P loading and carefully administered to maintain water column pH at 6 or above (Kennedy and Cooke, 1982). Alum treatment would also be most effective in areas of sediment accumulation (depth greater than  $2.6 \text{ m}$ ) of lakes and reservoirs (James et al., 2000). In this study, a  $0.25\text{-g}$  alum dose to the cores ( $79 \text{ g m}^{-2}$  or an estimated  $>900\text{-Mg}$  reservoir treatment) resulted in sediment P flux that was eight times less under anaerobic conditions. This dose was based on a 1-yr treatment strategy and the addition of alum should be much greater for long-term reservoir restoration plans. Several studies have observed significant improvement in water quality following alum treatment to bottom sediments (e.g., see Kennedy et al., 1987; Welch and Schrieve, 1994).

## Conclusions

This study demonstrated that sediment P flux was fourfold greater under anaerobic conditions compared with aerobic conditions, and that the internal P load from sediments was approximately 25% of the annual P load estimated from external sources. Thus, comprehensive watershed and reservoir P management plans in this catchment must consider internal sources, as well

as external sources. If internal sources in this reservoir are left unabated, then improved water quality may not be the result of the P-based management strategies currently being implemented on pastures and row crop areas in this watershed. A more detailed assessment of sediment P release should be considered in Lake Eucha, especially in areas of sediment deposition and accumulation.

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